Positive outcomes from U.S. lead regulations, continued challenges, and lessons learned for regulating emerging contaminants

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Short research and discussion article

Abstract

Albeit slow and not without its challenges, lead (Pb) emissions and sources in the United States (U.S.) have decreased immensely over the past several decades. Despite the prevalence of childhood Pb poisoning throughout the 20th century, most U.S. children born in the last two decades are significantly better off than their predecessors in regards to Pb exposure. However, this is not equal across demographic groups and challenges remain. Modern atmospheric emissions of Pb in the U.S. are nearly negligible since the banning of leaded gasoline in vehicles and regulatory controls on Pb smelting plants and refineries. This is evident in the rapid decrease of atmospheric Pb concentrations across the U.S over the last four decades. One of the most significant remaining contributors to air Pb is aviation gasoline (avgas), which is minor compared to former Pb emissions. However, continual exposure risks to Pb exist in older homes and urban centers, where leaded paint and/or historically contaminated soils+dusts can still harm children. Thus, while effective in eliminating nearly all primary sources of Pb in the environment, the slow rate of U.S. Pb regulation has led to legacy, secondary sources of Pb in the environment. More proactive planning, communication, and research of commonly used emerging contaminants of concern that can persist in the environment long after their initial use (i.e., PFAS) should be prioritized so that the same mistakes are not made again.

Keywords: Lead (Pb), emerging contaminants, environmental policy, environmental pollution

Introduction

While there are many different environmental contaminants of concern, one of the most publicized and widespread persistent contaminants in the global environment over the last century is lead (Pb). Lead is a metal on the periodic table with a high atomic weight (207.2), is soft, malleable, has a low melting point (327 $^{\circ}$ C), and is ductile—making it very useful for a variety of applications. These applications include use in gasoline, paints, batteries, and even fishing weights. However, Pb is also a severe neurotoxicant that particularly harms

children, with no known safe levels of exposure (e.g., Betts, 2012). Exposure to Pb is mostly due to ingestion pathways (e.g., Kranz et al., 2004, Moya and Phillips, 2014), although inhalation pathways from small particulate matter can also occur (e.g., Laidlaw et al., 2017). While this ingestion may be of the actual Pb-tainted object itself (i.e., Pb paint chips, toys, etc.), it is commonly via Pb-contaminated soil and dusts (e.g., Lanphear et al. 2002), particularly in modern cases of chronic Pb exposure.

Regulations both in the United States (U.S.) and globally have sought to eliminate major sources of Pb to the environment, particularly leaded gasoline and leaded paint. While oftentimes enacted slowly and inconsistently across countries, the curbing of primary Pb emissions has significantly decreased blood lead levels (BLLs) among many populations. For example, the elimination of leaded household paint in the U.S. in 1978 and final elimination of U.S. leaded gasoline in 1996 contributed to a reduction in average BLLs from 12.8 g/dL in 1976-1980 (geometric mean, aged 1-74 years) to 0.82 g/dL in 2015-2016 (e.g., Dignam et al., 2019). While this trend is common in analogous, higher income countries, it unfortunately isn't always the case in lower- and middle-income countries, where higher relative exposure still persists, often because of significant lead sources that are not leaded gasoline and paint (e.g., Attina and Trasande, 2013; Ericson et al., 2021).

Lead is a fairly immobile element under most environmental conditions, meaning that it tends to stay put, with residence times in soils sometimes approaching hundreds to thousands of years (e.g., Miretzky and Fernandez-Cirelli, 2008). Thus, following emission into the environment, Pb in soils may pose a secondary exposure risk long after it was initially released, which is a reason for continual chronic Pb exposure in many locations to this day, even when phased out of products. Regulation and exposure prevention are therefore twofold—needing to focus not just on the primary pollutant sources of Pb, but the legacy, secondary pollutant sources as well. While the issue of primary pollutant sources can be addressed more easily through effective regulation, cleaning up the mess of legacy pollutants, particularly at a large-scale, can be quite the challenge.

Despite inconsistencies and challenges with policy implementation worldwide, the successes and impacts of Pb regulation thus far should not be overlooked. Here, we emphasize what good has come from Pb regulations, with a focus on the U.S., the challenges that remain—particularly from a secondary exposure perspective, and what we can learn moving forward that is applicable not only to Pb, but other emerging legacy contaminants of concern such as microplastics and Per- and Polyfluoroalkyl Substances (PFAS).

Discussion

The good regarding Pb regulation

While it may be easy to focus on all the negative outcomes and news surrounding Pb pollution and poisoning in the U.S. over the last several decades—the Flint, Michigan disaster is one that comes to many people's minds—both scientists

and regulators mustn't forget how powerful eventual health intervention was on reducing Pb exposure. While it took decades and heavy advocacy against the Pb industry from scientists such as Clair Patterson and Howard Mielke to bring forth U.S. regulation prohibiting the sale of products made with Pb, the subsequent effects following these regulations is astonishing. Not only did BLLs in children and adults drop precipitously [a 93.6% drop from 1976-1980 to 2015-2016 in the U.S. (e.g., Dignam et al., 2019) and a 94.5% drop in U.S. children aged 1-5 from 1976-1980 to 2011-2016 (e.g., Egan et al., 2021)], but environmental measures of Pb emissions in air dropped immensely as well. For example, the concentration of median annual total suspended particulate (TSP) Pb from U.S. EPA air monitoring stations dropped nearly 99% from 1980 to 2016 (Fig. 1). Most of the decline occurred in the 1980s, in conjunction with the rapid phaseout of leaded gasoline in the U.S. (Fig. 1A). Following the complete ban of leaded gasoline for on-road vehicles in the U.S. via the Clean Air Act in 1996, atmospheric Pb dropped even more. Over time, atmospheric Pb concentrations have decreased so significantly that most U.S. air monitoring stations have phased out analyzing for Pb.

With the decrease in atmospheric Pb, annual Pb input into soils and sediments has also drastically declined over time. Mielke et al. (2011) estimate that there was around 4.64 million metric tons (MT) of Pb gasoline additive consumption from 1950-1982 in the U.S., with about 75% of this emitted into the atmosphere. Additionally, the authors state this was about 86% of the total Pb gasoline additive consumption from 1927 to 1994. First order estimates of Pb atmospheric loading can be made using EPA Pb TSP median values by decade and assuming 85 m^3 of air filtered per day by air monitor, with an air filter surface area of 0.05 m^2 [assumptions explained in Dietrich (2020)] (Fig. 2A). Since 1980, the estimated annual deposition of Pb in MT has decreased substantially by decade (Fig. 2A), and even in the 1980s the estimated mass of atmospheric Pb was 75,780 MT, or only 1.6% of the estimated Pb gasoline additive consumption from 1950-1982. Approximations of Pb deposition effects on Pb concentrations in surface soils (upper 5 cm) can be made by (1) using Pb air deposition estimates, assuming an average soil density of 1.7 g/cm^3, (2) assuming that Pb deposition was primarily concentrated in urban areas from leaded gasoline and industry, and (3) urban square area estimates by decade as ascribed by the U.S. Bureau of the Census (their definition of urban areas). In the 1980s, it is estimated that atmospheric Pb deposition in urban areas on average raised soil Pb concentrations by 11.7 mg/kg, but by 2010, the effect was essentially negligible (Fig. 2A).

This decrease in atmospheric pollutants wasn't limited to Pb, but extends to other pollutants as well, such as SO_2—a harmful gas that is particularly known for contributing to acid rain. Median SO_2 values at air monitoring sites throughout the U.S. have drastically decreased over the last several decades, largely due to the U.S. EPA's Acid Deposition Program and cleaner, more efficient processes such as flue-gas desulfurization during fossil fuel burning (Fig. 3). However, atmospheric pollution, particularly in urban centers, remains a relatively sig-

nificant contributor to global mortality (Fig. 4). $PM_{2.5}$ and NO_2 atmospheric pollutants are pervasive in many parts of the world, where greater pollution regulation is needed to reduce human exposure (Health Effects Institute, 2020).

As of 2017, the primary new source for Pb in the U.S. environment is avgas—gasoline used for aviation single piston aircraft, comprising 70% of total Pb emissions (U.S. EPA, 2017). While any source of Pb to the environment is bad, the amount of Pb emissions annually projected from avgas is 470 tons (U.S. EPA, 2017), only 0.19% of the peak amount of Pb emissions, 250,000 tons, in the 1970s. Even with the relatively low amount of Pb emissions from avgas though, the high toxicity of Pb has led the U.S. EPA to recently issue a proposed "endangerment finding" due to concerns that avgas may still "reasonably be anticipated to endanger public health and welfare under section 231(a) of the Clean Air Act" (U.S. EPA, 2022b). Following an official endangerment finding in 2023, the EPA would then propose regulatory standards from aircraft.

In addition to drastically less atmospheric Pb emissions over the last 40 years, the elimination of Pb paint in residential housing in 1978 has led to much less household Pb exposure in many parts of the U.S. However, despite the clear decreases in Pb sources and exposure over time, hundreds of thousands of children in the U.S. are still exposed to unnecessary amounts of Pb from historic Pb sources, largely associated with risk factors such as poverty, race/ethnicity, and housing age (e.g., Egan et al., 2021).

Lingering Pb

Most of the chronic Pb exposure that occurs today in the U.S. (and many other countries) is due to childhood ingestion of Pb—with soil and dust being a major component of that. However, it is acknowledged that inhalation of airborne particulate Pb is an often underappreciated exposure source (e.g., Laidlaw et al., 2017). Two of the major sources of Pb in soils and dusts are from historic leaded gasoline and leaded paint, the legacy of both remaining printed on soils and houses decades after regulations eliminated their use. Urban settings in particular are replete with these soil and dust repositories of contamination, posing a continual health risk to children via ingestion during hand-to-mouth activities (e.g., Mielke and Reagan, 1998; Mielke et al., 2007). While regulation in the U.S. has effectively limited the amount of "new" Pb being set forth into the environment, there has been inconsistent regulation on what to do with secondary sources of Pb in the environment, such as Pb-tainted soils and dusts.

The current U.S. EPA residential soil standard of 400 mg/kg for Pb has been criticized for being outdated, inconsistent with dust Pb loading regulations, and not providing an adequate margin of safety for children's health (e.g., Gailey et al., 2020; Laidlaw et al., 2017). The steepest increase in children's BLLs occurs at the lowest soil Pb concentrations (e.g., < 100 mg/kg; Mielke et al., 2007) and the greatest effects of Pb exposure on intellectual impairment are at lower BLLs (e.g., Lanphear et al., 2005). To fully eradicate childhood Pb poisoning at any level in the U.S., it is essential to develop consistent and effective Pb guidelines

for soils and dusts, particularly to keep in line with the recent lowering of the CDC BLL guidance from 5 µg/dL to 3.5 µg/dL in children.

Part of the impetus necessary to promote awareness of health risks associated with Pb in soils and dusts and the associated regulatory changes needed is a shift in public perception on lead risks. With the national media coverage and publicization of the Flint, MI water crisis and large push from the recent U.S. Bipartisan Infrastructure Law to replace lead service lines—many Americans perceive water as the biggest threat for Pb exposure. In fact, in our experience with multiple community science programs providing free Pb testing, the type of environmental media we are asked most often about is water. While water is indeed a potential Pb exposure risk and all lead service lines should eventually be replaced so no threat of Pb exists, water is presently not the main exposure route of Pb for most children. Take for example, Flint, MI, where the switch to Flint River Water and subsequent corrosion of lead piping dramatically increased drinking water Pb concentrations. Incidence of elevated blood Pb levels (EBLLs, 5 µg/dL) in children doubled immediately after the incident (2.4% to 4.9% ($P < .05$); Hanna-Attisha et al., 2016), but many children were already exposed to harmful levels of Pb beforehand. Even in 2010, EBLLs were at rates as high as post-incident levels in 2014-2015 (Laidlaw et al., 2016). Based on the seasonality of children's (< 6 years old) BLL concentrations in Flint, elevated concentrations of Pb in Flint inner-city soils, and similarities to seasonal BLLs and atmospheric Pb trends in other cities, Laidlaw et al. (2016) contends that soil+dust are indeed an important source of Pb exposure in Flint.

While other cities that have had/have water corrosivity problems in lead piping, resulting in water Pb concentrations spikes (e.g., Washington D.C. in the early 2000s), many cities with lead service lines have water Pb concentrations well below EPA guidelines. For example, tap water samples in New Orleans, LA from residents under normal use conditions in 2015–2017 (n = 1497) and under a variety of methods (i.e., first draw cold sample, 5.5-6 min cold water flush, first draw hot water sample, etc.) had a 90^{th} percentile water Pb level of 5.6 ppb, well below the EPA Action Level of 15 ppb (Katner et al., 2018). While many more samples (60%) exceeded the more stringent American Academy of Pediatrics recommended water lead level for schools (1 ppb) and obviously any concentration of Pb in water is not ideal, recent research on lead exposure modeling in a typical U.S. urban center (Milwaukee, WI) revealed that exposure risk to Pb is much more significant from soil+dust and foods grown in Pb-rich urban soils than Pb in water (Byers et al., 2020). The authors even demonstrated that when using soil+dust Pb values of 400 ppm, garden produce Pb values of 0.4 ppm, and the 90^{th} percentile of Flint, MI water values (26.8 ppb) from August of 2015 (before switching back to treated Lake Huron water in October of 2015), that water still only contributed to <15% of total estimated child Pb ingestion. This trend holds up internationally as well, where Pb isotope analyses of children's blood in France from 2008-2009 (representative of children aged 6 months to 6 years old with a blood lead level 25 g/L) revealed that when a single source of Pb could be identified, it was only from water 5%

of the time ($CI_{95\%}$ = 0-11%) (Oulhote, 2011). Thus, while all sources of Pb are important to consider from a health intervention standpoint, the question arises as to whether it is better to initially focus on Pb soil+dust remediation to reduce child BLLs over lead service line replacement, which is extremely costly (anywhere between 20-50 billion USD for the entire U.S.) and can potentially re-contaminate the water supply with Pb during replacement.

Suggestions moving forward—Pb and beyond

Many compelling reasons exist to lower the U.S. national EPA soil Pb safety standard of 400 ppm for residential properties (Lupolt et al., 2021; Saikawa and Filippelli, 2021), particularly because this was based on the soil Pb-BLL relationship defined decades ago for a BLL limit of 10 µg/dL. Now that the BLL standard has been reduced, the soil Pb standard should follow suit. Two issues remain with finding the "right" soil Pb standard value—the relationship between soil Pb and BLL is very poorly constrained below 400 ppm soil Pb, and the sheer number of urban properties with soil Pb above 400 ppm is tremendous. Even more urban areas would be eligible for U.S. EPA Superfund classification if the 80 ppm residential soil Pb standard in California were to be adopted nationally. For example, in 2019, the median soil Pb concentration in Detroit, MI urban residential sites was still 92 mg/kg, meaning at least half of the areas sampled would be over the 80 mg/kg standard (Mielke et al., 2020). The procedure of characterizing these urban areas, litigating past polluters from different sources and related industries—including leaded gasoline, Pb-based paints, and industrial emissions—and conducting extensive soil remediation actions across many major U.S. cities would be staggeringly expensive. Yet, something must be done about it, as soil (collectively with the dust it generates) is a higher exposure risk source than water systems, which are receiving tens of billions of dollars for replacement. Not to get us wrong here—every dollar used to eliminate sources of Pb is a dollar well spent, but a greater return on investment will come from addressing the urban soil Pb repository issue.

While admittingly more still needs to be done regarding cleaning up Pb in the environment to prevent exposure, some clear lessons have been learned that can be applied to other emerging contaminants of concern in the environment:

1. **Adopt the precautionary principle more frequently whenever dealing with chemicals/elements whose structures or formulas have limited toxicological research, but pose the potential to persist in the environment and accumulate in organisms.**
    (a) The basis beyond Pb regulations was not based on the precautionary principle, but instead placed the burden of proof on researchers to prove that the use and exposure to Pb was detrimental beyond the economic benefit of use. This reactionary approach has ended up costing people far more in the long run than any benefits provided by Pb-based products, such as tetraethyl lead.
2. **Incentivize collaborative corporate and academic research on the**

potential health and environmental implications of products, instead of creating silos of research.

   (a) Like governmental market-based initiative programs such as the Acid Rain Program that have been successful, funding and support for collaborative industrial research on environmental fate and toxicity of products would likely improve companies' abilities to evaluate risk and harm of their products. This would also provide outside, objective researchers a chance to evaluate the data, and potentially save millions in taxpayer dollars down the road if potential emerging contaminants of concern had not been caught in the production phase.

3. **Implement consistent methods and effective communication of toxicological/health exposure studies to inform effective legislation, particularly for secondary sources of pollutant exposure from water, soil, and air.**

   (a) While Pb has been studied more than most toxic substances, unclear communication, inconsistent study methodology, and inconsistent evaluations of risk have still made regulatory policies in some environmental media difficult to enact or ineffective. Specifically, regulations for Pb in dust do not match Pb in soil regulations, nor have there been any recent updates in these regulations to keep pace with secondary prevention of children with EBLLs (i.e., the changing CDC blood Pb guidelines for intervention). Additionally, there needs to be better public communication of main risks of exposure—such as with Pb in soil+dust—to give people individual agency in exposure reduction.

Unfortunately, these lessons will likely need to be applied to a growing plethora of emerging contaminants in the environment. While there are many, two types of contaminants with much recent concern are the persistent PFAS "forever" chemicals and microplastics. Both of these can persist for long periods of time in the environment and travel long distances—as far as polar ice sheets. While we as a society have started using both PFAS and microplastics in products extensively over the last several decades, we know remarkably little about their actual environmental health impacts and how they react over time under different environmental conditions. For instance, there are thousands of PFAS chemicals, but most studies have only focused on the toxicity of a limited number of them.

Not all is gloom and doom with how we can approach these new environmental challenges. It is clear through research and policy enactment with previous contaminants in the environment that we are capable of addressing pressing issues of both legacy contaminants and new contaminants of concern. We need to learn from our past mistakes and begin to value long-term collective environmental and human health over any short-term benefits gained from new market products to prevent similar environmental health disasters that have occurred

in the past.

Declarations

Ethics approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

Availability of data and materials

The datasets generated and/or analyzed during the current study are available at: https://aqs.epa.gov/aqsweb/airdata/download_files.html#Daily

Competing interests

The authors declare that they have no competing interests.

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Authors' contributions

MD: Original draft writing, conceptualization, data compilation, figure generation.

GMF: Review and editing, conceptualization.

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Figure captions

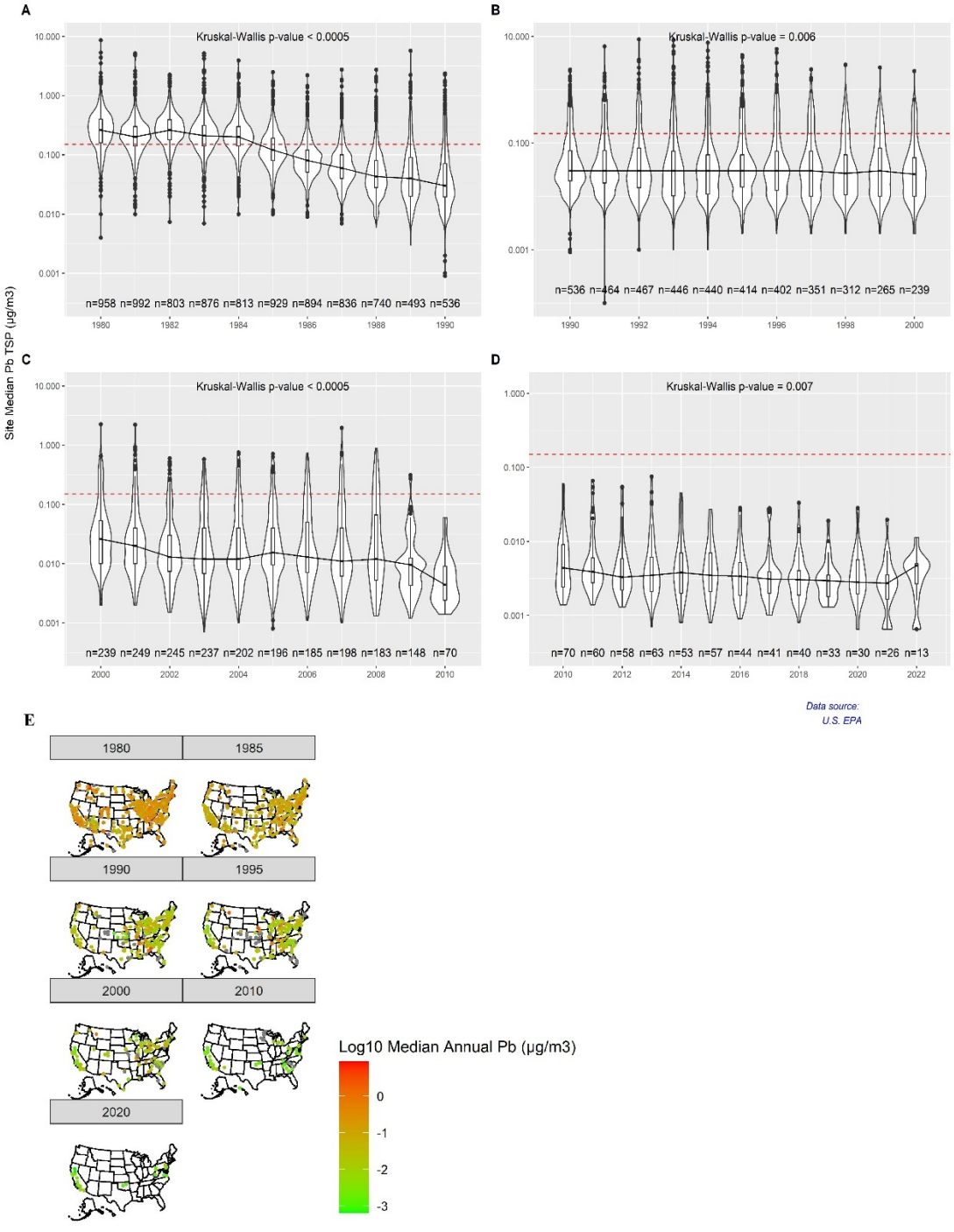


Figure 1: (A-D) Median annual Pb total suspended particulate matter (TSP)

values (including 1-month, composite, and 24-hr. data) per air monitoring site in the U.S. in µg/cm^3 (log10 transformed y-axis), with panels A-D grouped by decade. The boxes represent the interquartile range (IQR) of 25–75 percentiles of data, the horizontal line within and between the boxes represents the median, and the whiskers represent 1.5 times the IQR. The dotted red line is NAAQS standard of 0.15 µg/cm^3 for a three-month period. Non-parametric Kruskal-Wallace test p-values are also provided. (E) Maps with log10 transformed values of median annual Pb concentrations throughout the U.S. shown in color. Grey samples are those that had a raw Pb concentration of 0 µg/cm^3. Notice the decreasing Pb TSP values over time but also the decrease in Pb air monitoring sites. Source: U.S. EPA (2022a).

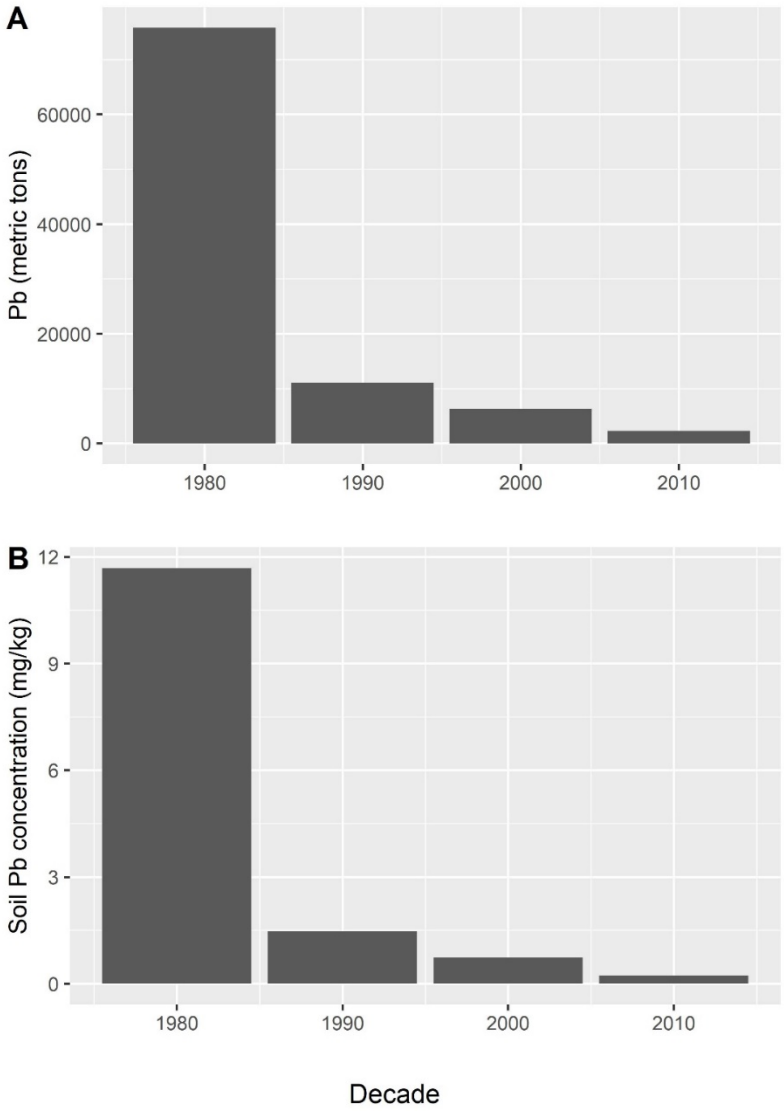
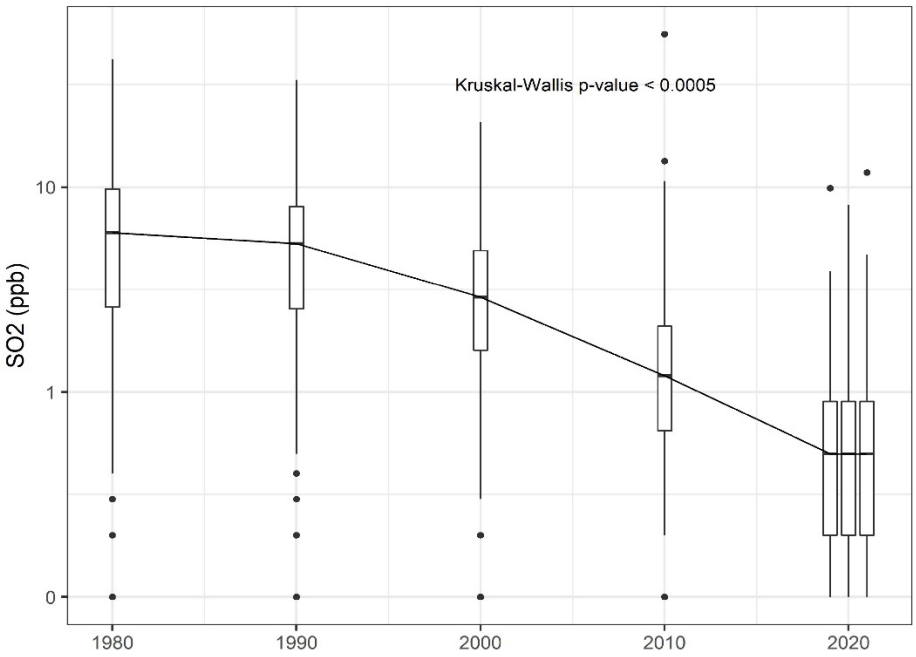


Figure 2: (A) Estimated atmospheric Pb deposited per decade in metric tons in urban areas within the U.S. (B) How much soil Pb concentrations are estimated to have increased in urban areas throughout the U.S. by decade from continuous atmospheric Pb deposition. This assumes Pb is not lost from the soil, although there are likely urban soil resuspension processes reworking some of this Pb contaminated soil (e.g., Laidlaw and Filippelli, 2008).

A Annual median SO2 air measurements



B

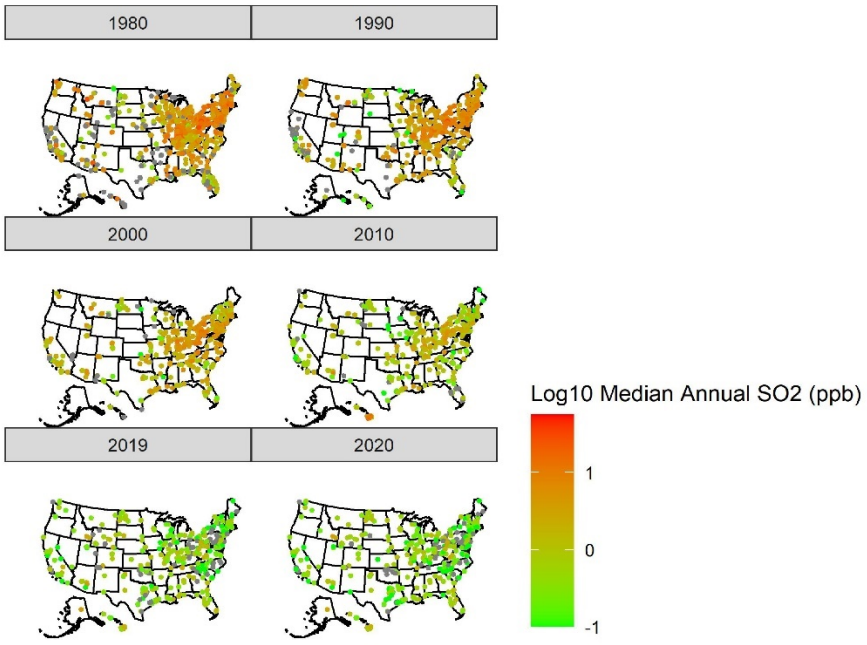


Figure 3: (A) Median annual SO_2 values (24 hr) per air monitoring site in the U.S. in ppb (log10 transformed y-axis). The boxes represent the interquartile range (IQR) of 25–75 percentiles of data, the horizontal line within and between the boxes represents the median, and the whiskers represent 1.5 times the IQR. A non-parametric Kruskal-Wallace test p-value of <0.0005 demonstrates that there is a high likelihood at least one population SO_2 median value differs from the other populations (years). (B) Maps with log10 transformed values of median annual SO_2 concentrations throughout the U.S. shown in color. Grey samples are those that had a raw SO_2 concentration of 0 ppb. Notice the decreasing SO_2 values over time. Source: U.S. EPA (2022a).

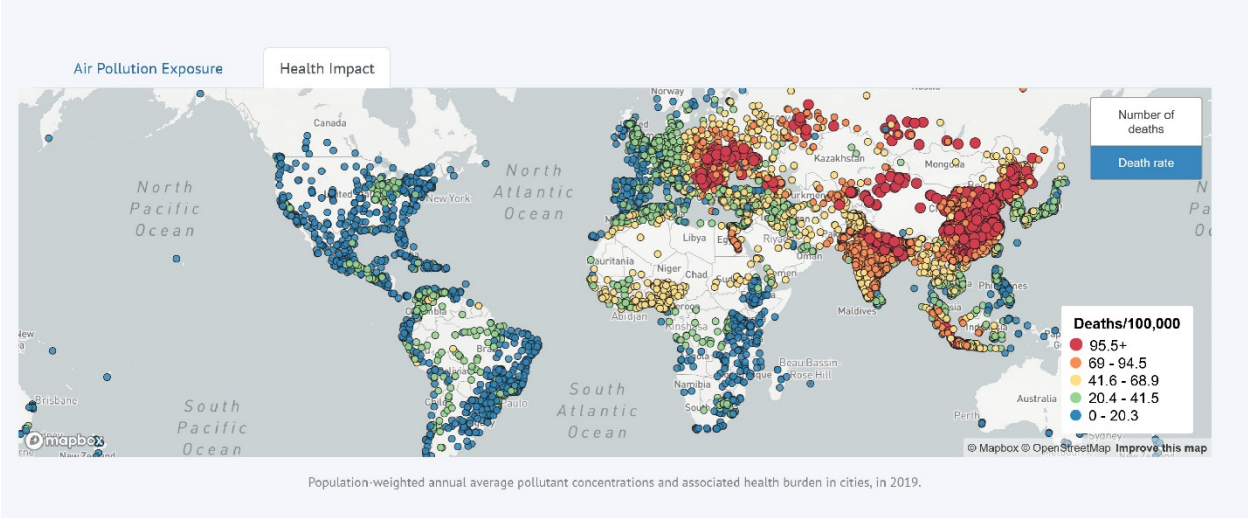


Figure 4: Screenshot from the State of Global Air 2020 report (Health Effects Institute, 2020: https://www.stateofglobalair.org/resources/health-in-cities#exposure_to_no_sub_2_sub_) displaying the projected deaths per 100,000 people ascribed to air pollution exposure in cities in 2019.